

LA-UR- 03-0474

Approved for public release;
distribution is unlimited.

Title: Heat Removal from High Temperature Tubular Solid Oxide
Fuel Cells Utilizing Product Gas from Coal Gasifiers

Author(s): William Jerry Parkinson

Submitted to: 28th International Technical Conference on Coal Utilization &
Fuel Sytstems
March 10-13, 2003
Clearwater, Florida



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Form 836 (8/00)



Heat Removal from High Temperature Tubular Solid Oxide Fuel Cells Utilizing Product Gas from Coal Gasifiers

William J. Parkinson
Los Alamos National Laboratory
Los Alamos New Mexico

ABSTRACT

In this work we describe the results of a computer study used to investigate the practicality of several heat exchanger configurations that could be used to extract heat from tubular solid oxide fuel cells (SOFCs). Two SOFC feed gas compositions were used in this study. They represent product gases from two different coal gasifier designs from the Zero Emission Coal study at Los Alamos National Laboratory. Both plant designs rely on the efficient use of the heat produced by the SOFCs. Both feed streams are relatively rich in hydrogen with a very small hydrocarbon content. One feed stream has a significant carbon monoxide content with a bit less hydrogen. Since neither stream has a significant hydrocarbon content, the common use of the endothermic reforming reaction to reduce the process heat is not possible for these feed streams.

The process, the method, the computer code, and the results are presented as well as a discussion of the pros and cons of each configuration for each process.

INTRODUCTION

The motivation for this study was the need to determine if the heat generated as a by-product of electrical power generation from a fuel cell could be captured and used for endothermic process requirements elsewhere in our plants. The Los Alamos Zero Emission Coal team is currently looking at two different coal gasification schemes to provide hydrogen rich fuel feed for fuel cells that produce electrical power and heat. Both of these plants have been designed using ASPEN Plus®, a state-of-the-art plant simulation computer code. Both ASPEN Plus® simulations are completely workable from a purely thermodynamic design basis. That is based upon chemical equilibrium calculations, mass and energy balances, these simulations provide compatible unit operations with reasonably large temperature differences for assumed heat transfer. This study was initiated to determine if equipment configurations, stream flows, and physical properties could be manipulated in a way to provide heat transfer coefficients that could actually provide the assumed heat transfer. A future study will be needed to determine if the equipment specified in the heat transfer study can actually be fabricated, before we can be fully satisfied with our plant designs.

Although ASPEN Plus® has some heat transfer capabilities, they were not adequate for this study. Therefore we had to write our own heat transfer computer codes. We wrote two codes. One was a basic shell-and-tube heat exchanger code and is strictly for turbulent flow. This code allowed the user to vary tube size and pitch easily along with the other important heat transfer parameters. This code was used to determine the heat transfer requirements of the more standard unit operations. The second code, the one discussed here, allows for flexible configurations, but utilizes the same physical property information as the first code. This code also has some chemical reaction capability and allows for laminar flow heat transfer. This is the code that we used to analyze the fuel cell heat transfer capabilities.

THE PROBLEM

The two different coal gasification schemes under study at the Los Alamos National Laboratory are different. Both require the heat energy input from the fuel cell, but in different plant locations at different operating temperatures. Therefore we had to do a separate study for each plant design. Figures 1 and 2 are simplified flow diagrams of the portions of the two schemes that are of interest in this study. Figure 1 is from the original Los Alamos coal gasification scheme [1] that uses an exothermic hydro-gasification step, a carbonation step that produces hydrogen from the gasifier product and calcium oxide, and an endothermic calcinations step that produces calcium oxide from calcium carbonate. Figure 2 is a simplified gasification scheme that eliminates the carbonation and calcinations steps but utilizes an endothermic gasifier.

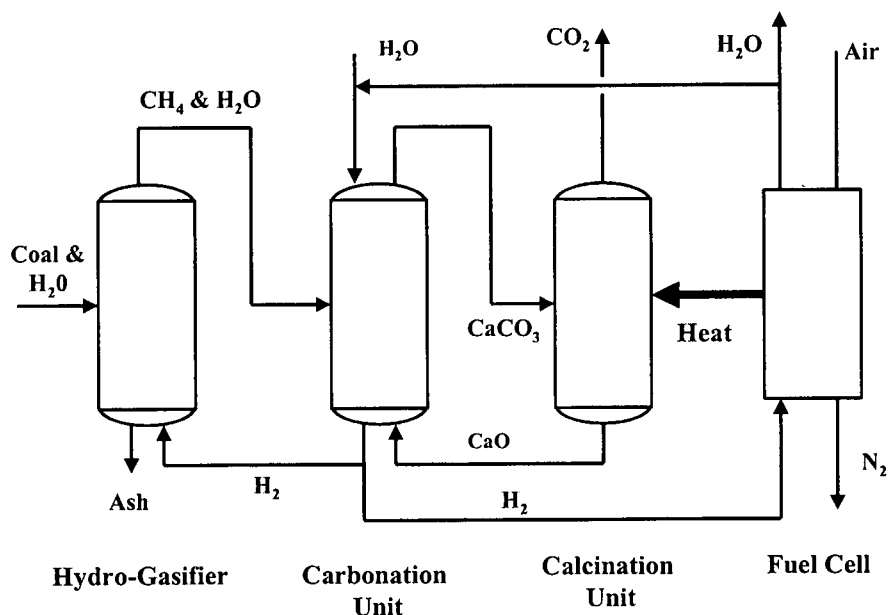


Figure 1. Simplified schematic of the exothermic hydro-gasification process.

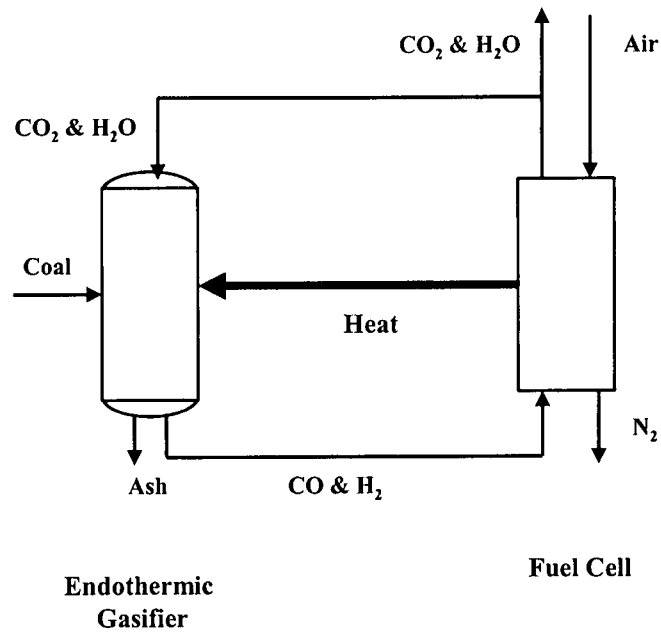


Figure 2. Simplified schematic of the endothermic gasification process.

The composition, temperature and pressure of the fuel gas supplied to the fuel cell for both Case 1, Figure 1, and Case 2, Figure 2, are given in Table I.

Table I. Fuel Cell Feed Streams for Both Case 1 and Case 2.

Mole Fraction	Case 1 –Exothermic Hydro-Gasification	Case 2- Endothermic Gasification
H ₂ O	0.5428	0.0675
N ₂	0.0066	0.0046
CO	0.0085	0.5592
H ₂	0.4263	0.2225
CO ₂	0.0111	0.1443
CH ₄	0.0047	0.0019
O ₂	0.0	0.0
Temperature (°F)	2192	1800
Pressure (psi)	424	60

In each case, the heat transfer target is operated at different temperatures and the fuel cells are operated at differently. In Case 1, the heat transfer target, the calciner, is operated at 1679 °F. In Case 2, the heat transfer target, the gasifier, is operated nominally at 1600 °F. The operation of the fuel cells for each case is discussed in more detail in the section describing the computer study and the results of the computer study.

THE FUEL CELL

The fuel cell model chosen for this study was based on the tubular solid oxide fuel cell design by Siemens Westinghouse Corporation, discussed in reference [2]. Some information was also obtained from the Siemens Westinghouse website. We chose this model because, of those currently available, it appears to most nearly satisfy our plant needs of high temperature and pressure. The model used in this study was not an exact replica of the Siemens Westinghouse model for two reasons. One, we don't have all of the proprietary information about the fuel cell to model it exactly. Two, we assume that improvements and changes to this fuel cell will be made prior to our need for it.

The fuel cell is shown in Figures 3 and 4. As shown in Figure 3, the cell is built in layers. The layers consist of an air cathode, and electrolyte layer, and a fuel anode. The cells are connected in series and are packed in bundles much like a shell-and-tube heat exchanger as shown in Figure 4. Each cell or tube has a closed end and the air is injected using a delivery tube as shown in Figure 3. For our model each tube is 150 cm in length with a 2.2 cm nominal diameter.

Typically the feed to these fuel cells is natural gas. This natural gas is reformed at the fuel cell inlet to provide a hydrogen rich fuel for the oxidation reaction. The reforming step is endothermic, providing a heat sink for the exothermic oxidation reaction. Our situation is different. We provide the fuel cell with the oxidation reactants directly and we

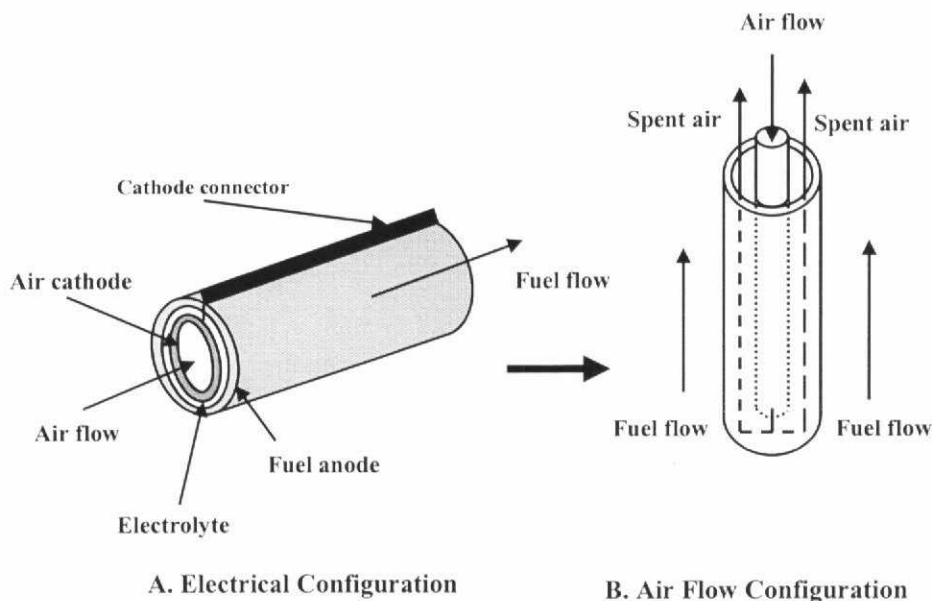


Figure 3. Configurations for a single tube from the solid oxide fuel cell.

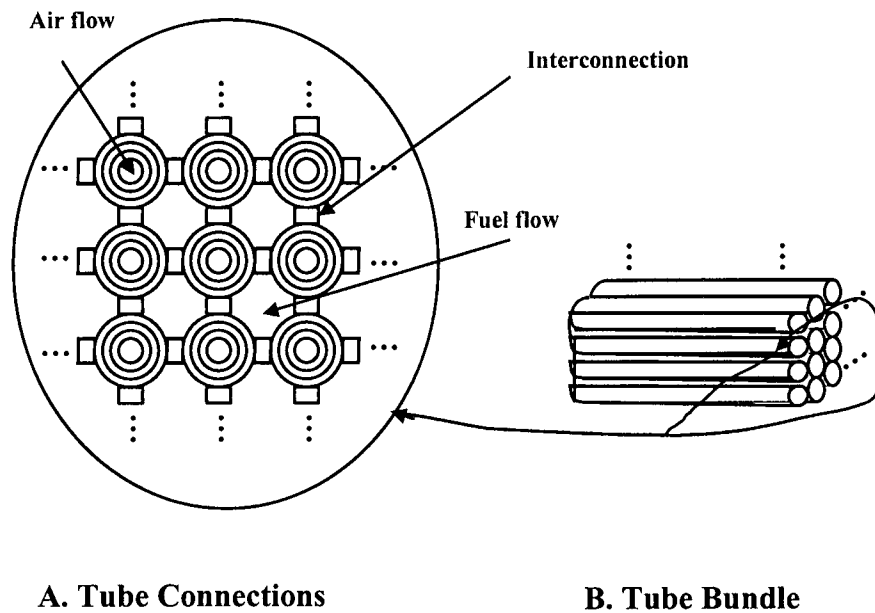


Figure 4. Tube bundle configuration for the solid oxide fuel cell.

want to capture the energy from the exothermic reaction to use elsewhere in our plants. In this study we look at three different configurations for removing heat from the fuel cell. They are shown in Figure 5. Configuration I, shown in Figure 5A, adds an extra tube around each fuel cell to provide an annulus to contain the fuel flow. The coolant then flows parallel to the tube bundle as the fuel does in the original design. This configuration may prove difficult to fabricate because of the cathode-anode connection required. Fabrication problems will be explored in a future study as shown in Figure 6. Configuration II, shown in Figure 5B, adds separated tubes for coolant flow to the original fuel cell design. Configuration III, Figure 5C, utilizes normal fuel cell operation and then removes the heat of reaction from the spent fuel stream with a standard shell-and-tube heat exchanger. This configuration was not tested with the exothermic hydro-gasification process because the simulation requires the energy to be removed inside the fuel cell.

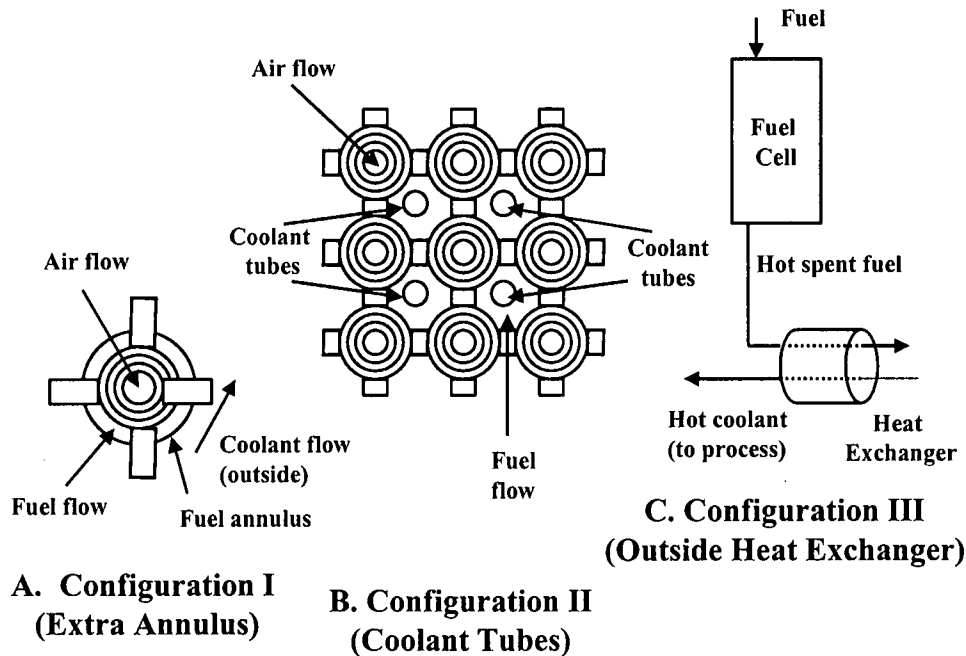


Figure 5. Heat exchange configurations for fuel cell study.

The basic fuel cell assumptions used in this study were:

- Fifty percent of the exothermic energy generated from the oxidation reaction becomes electrical energy and the other fifty percent becomes heat.
- The electrical power produced by each cell is 0.2 watts/cm^2 of active surface area.
- Eighty five percent of the fuel is utilized.
- Twenty five percent of the oxygen in the air is utilized.
- Since we don't know the reaction kinetics of these systems, we assume that the space velocity for the fuel must be approximately the same as that of the normal fuel cell in order to maintain the 85 percent conversion.

The physical properties used in the model were obtained from references [3,4].

This portion of the overall study, the transport problem is only a portion of the problem. It has to be considered in prospective with the process design and the fabrication design. This is often an iterative process. Figure 6 illustrates this point.

THE COMPUTER CODE

The computer code developed for this study relies heavily on physical properties of the fluid streams to determine the heat transfer coefficients. Nearly all convective heat transfer coefficient correlations, both for laminar and turbulent flow, are some function of the fluid thermal conductivity, Reynolds number, and Prandtl number, as shown in Eq 1.

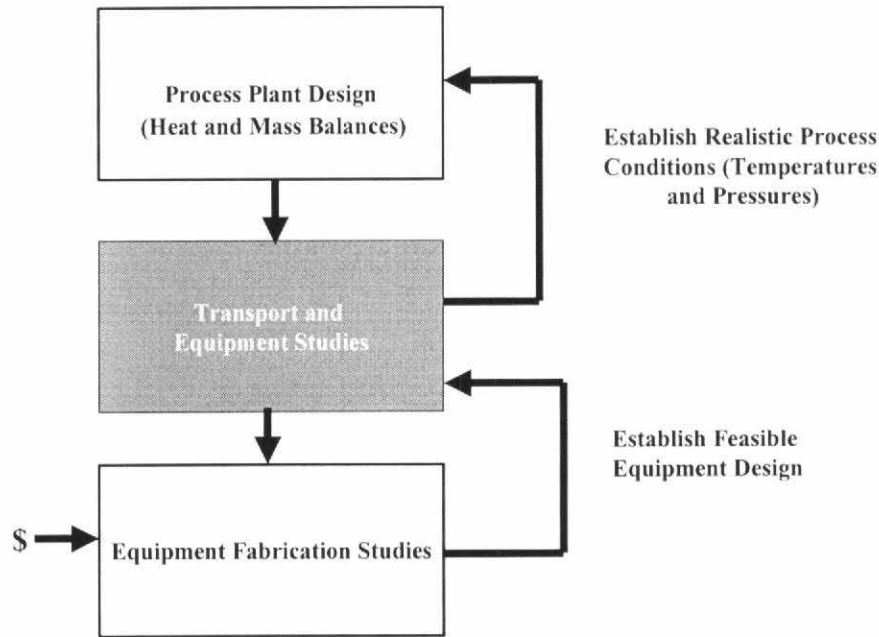


Figure 6. The role of the transport study in the overall scheme.

$$h = f(k_f, N Re, N Pr) \quad (1)$$

Where h is the heat transfer coefficient and k_f is the fluid thermal conductivity. NRe is the Reynolds number equal to $VD\rho/\mu$ and NPr is the Prandtl number equal to $C_p\mu/k_f$. Here V is the fluid velocity, D is the diameter or equivalent diameter of the conduit through which the fluid is flowing. ρ is the fluid density, μ is the fluid viscosity, and C_p is the fluid heat capacity. It is important to note here that most of the fluids that we deal with in this problem are mixtures. Fortunately, in our case, the mixtures are gases. Gases at the temperatures and pressures of these problems can be considered ideal mixtures. This means that the density and the heat capacity of these mixtures are essentially weighted averages of the individual components. This is not true, however, for the transport properties, viscosity and thermal conductivity. For mixture viscosities we use the Wilke mixing rule [5] and for the mixture thermal conductivities we use the Wassiljewa mixing rule with the Mason-Saxena modification [5]. For the pressures considered in this study, the affect of pressure is very small on the transport properties. So in this code the only pressure dependent property is the density.

In this code a reasonable amount of effort is devoted to computing the mixture physical properties as a function of temperature and composition, and in the case of density, pressure. In most heat transfer calculations temperature varies over the length of the tube, so the physical properties change and so do the heat transfer coefficients. In this problem, due to chemical reactions, the composition of the fuel stream and the air stream also vary with the length of the tube, so the heat transfer coefficient can vary even more with the

length of the tube. Therefore, it is important to know the stream compositions at all points in the heat exchanger. It is also important to be able to break the heat transfer zones into smaller increments to compute more accurate heat transfer.

In a typical heat exchanger, heat is transferred from one fluid, through a tube wall, into another fluid. This requires an overall heat transfer coefficient utilizing heat transfer coefficients calculated from equations of the form of Eq. 1 and incorporating the resistance to heat transfer offered by the tube wall. The overall heat transfer coefficient is determined by Eq. 2. The nomenclature is shown in Figure 7.

$$U_o = \frac{1}{\frac{1}{h_o} + R_o + R_k + \frac{R_i A_o}{A_i} + \frac{A_o}{h_i A_i}} \quad (2)$$

Where U_o is the overall heat transfer coefficient based on the outside tube area, and h_o and h_i are the heat transfer coefficients for the outside and inside streams, respectively. A_o , A_i , and R_o and R_i are the surface areas and scaling resistances for the outside and the inside of the tube respectively. R_k is the thermal resistance of the tube itself, defined by Eq. 3.

$$R_k = \frac{r_o \ln\left(\frac{A_o}{A_i}\right)}{k} \quad (3)$$

Where r_o is the inside radius of the tube and k is the thermal conductivity of the tube.

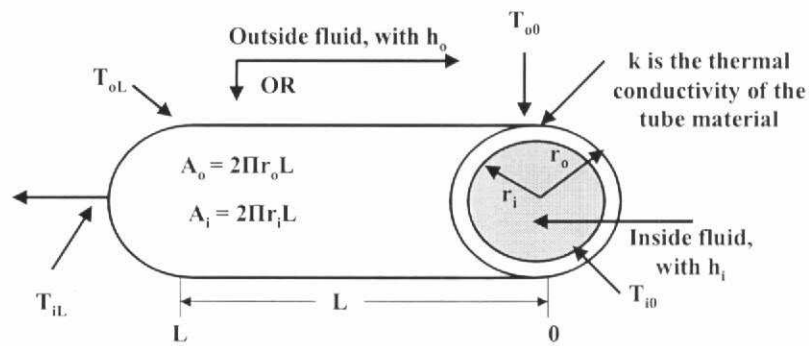


Figure 7. Heat transfer nomenclature

The computer code has several scale resistance types built into it, but for this study we assumed clean tubes and didn't use the factors R_o and R_i .

The heat transfer, Q , is defined by Eq. 4.

$$Q = U_o A_o \Delta T_{l.m.} \quad (4)$$

Where $\Delta T_{l.m.}$ is the log-mean temperature difference defined by Eq. 5.

$$\Delta T_{l.m.} = \frac{\Delta T_0 - \Delta T_L}{\ln\left(\frac{\Delta T_0}{\Delta T_L}\right)} \quad (5)$$

Where ΔT_0 is the temperature difference between the inside and outside fluid at the 0 end of the tube, $T_{o0} - T_{i0}$. See Figure 7. ΔT_L is the temperature difference between the inside and outside fluid at the L end of the tube, $T_{oL} - T_{iL}$.

The fuel cell problem is more complex. For configurations I and II, three equation similar to Eq. 4 must be solved simultaneously. With configuration III, only two of these equations need to be solved simultaneously. A third heat transfer equation is then solved independently using a separate shell-and-tube heat transfer code. Figure 8 shows an example of the complexity of the heat transfer for configuration I (very similar for configuration II).

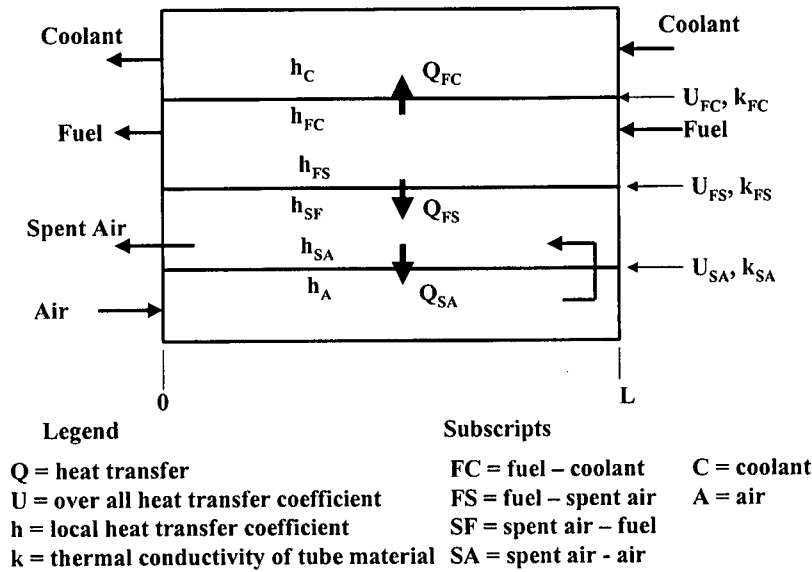


Figure 8. Heat transfer in the fuel cell, configuration I.

Note that in the case of the fuel tube and the spent air tube that two local heat transfer coefficients are computed for each tube, h_{FC} and h_{FS} for the fuel tube and h_{SF} and h_{AS} for the spent air tube. The difference is because the actual equation 1, used in the heat transfer calculations has a correction for the difference between the bulk fluid

temperature and/or viscosity and that of the fluid at the tubes surface temperature. In both of these cases, the tube surface temperature is different on the inside of the tube than it is on the outside of the tube. These are the local heat transfer coefficients used in Eq. 2 to compute the overall heat transfer coefficients. Another point to note is that all flows but the air flow are annular flows. For annular flow, the diameter used for the Reynolds number in Eq. 1 is the equivalent diameter. The equivalent diameter is defined as four times the flow cross-sectional area divided by the wetted perimeter. We describe the energy transfer to heat and cool the various streams by Eqs. 6-9.

Air:

$$Q_{air} = \overline{m}_{air} * \overline{C}_{Pair} * (T_{airL} - T_{air0}) \quad (6)$$

Where \overline{m} and \overline{C}_p with the over-bars refer to the average mass flow rate and average heat capacity, respectively.

Spent Air:

$$Q_{spa} = \overline{m}_{spa} * \overline{C}_{Pspa} * (T_{spa0} - T_{spaL}) \quad (7)$$

Where the subscript spa refers to the spent air stream.

Fuel:

$$Q_F = \overline{m}_F * \overline{C}_{PF} * (T_{F0} - T_{FL}) \quad (8)$$

Coolant:

$$Q_C = \overline{m}_C * \overline{C}_{PC} * (T_{C0} - T_{CL}) \quad (9)$$

If Q_R is the energy generated by the oxidation of the fuel, and half of that energy is converted to electrical power, then Eqs. 10-13 must be satisfied simultaneously before a workable configuration can be obtained.

$$\frac{1}{2} Q_R = Q_F + Q_{FC} + Q_{FS} \quad (10)$$

$$Q_C = Q_{FC} \quad (11)$$

$$Q_{spa} = Q_{FS} - Q_{SA} \quad (12)$$

$$Q_{air} = Q_{SA} \quad (13)$$

Where Q_{FC} , Q_{SA} , and Q_{FS} are solved implicitly using Eqs. 1-5.

The computer code itself has several sections. One section computes the mixture physical properties for each stream as a function of temperature, pressure, and composition. Another section computes the chemical equilibrium composition of the fuel stream and the oxygen required from the spent air stream in order to supply the energy produced by the specified active area of the fuel cell and the desired temperature. The new compositions of the fuel stream and the spent air stream are needed to compute heat transfer coefficients. Another section computes the heat transfer coefficients for each

stream. Finally the overall heat transfer coefficients are compute. Figure 9 is a schematic diagram of this computer code.

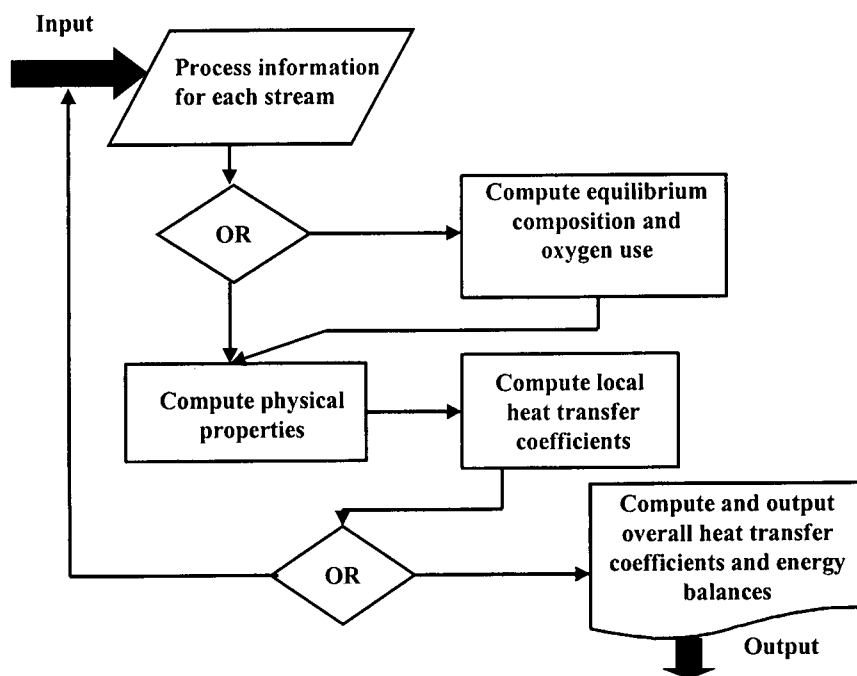


Figure 9. Block flow diagram for the fuel cell heat transfer code.

Upon closer observation, the reader will notice that the code is missing an outer loop. The code provides no mechanism to converge the energy balances given in Eqs. 10-13. This must be done by the user by trial-and-error. The procedure is to guess the temperature profiles for each stream. This requires four profile guesses for configurations I and II and three guess for configuration III. These guesses are input to the computer program. The profiles are updated for each successive run. There is also room to adjust the mass flow rate of the coolant and some room for changing the heat exchanger/fuel cell configuration. Convergence is achieved when Eqs. 10-13 are satisfied. If the user wants improved results, the exchanger/fuel cell can be cut in half and the above process repeated for each half. The procedure can be refined even further if desired. Figure 10. shows a typical temperature profile input guess for configuration I – case 2, the endothermic gasification process.

RESULTS

Five test cases were run. They were: configurations I and II for case 1, the exothermic hydro-gasification process, and configurations I – III for case 2, the endothermic gasification process. The information that is of interest for this study is the amount of heat transferred to the coolant from the fuel cell and the inlet and outlet temperatures of the coolant. These numbers are given in Table II for each of five cases studied.

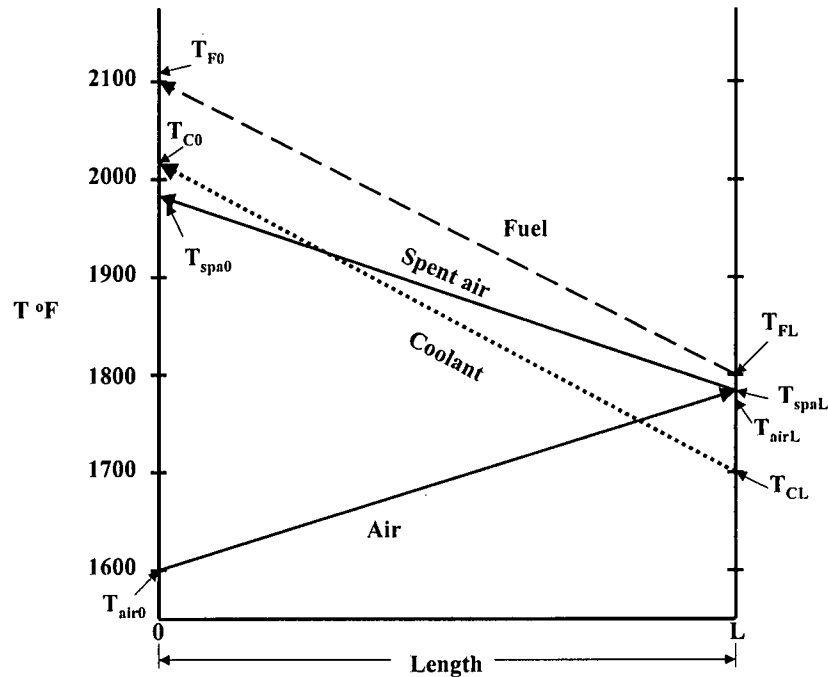


Figure 10. Typical input temperature profile guess for configuration I – case2.

Table II. Study Results -- Fuel Cell Heat Transfer for five Scenarios.

Scenario	Heat Transfer to Coolant/tube (Btu/ hr)	Coolant Inlet Temperature (°F)	Coolant Outlet Temperature (°F)
1- Case 1, Configuration I	505.3	1742	2050
2- Case 1, Configuration II	511.2	1735	2050
3- Case 2, Configuration I	480.7	1730	2008
4- Case 2, Configuration II	487.6	1723	2008
5- Case 2, Configuration III	503.1	1706	2008

For each scenario, the energy generated by the oxidation of the fuel, is adequately removed by the coolant. A portion of the energy generated, in each case is transferred to the spent air, approximately 25 percent. For each scenario, the energy generated and the coolant temperatures are close to those required by the process simulation. Each simulation will probably have to be adjusted slightly to allow for the heat transfer capability of the equipment. In addition, the feasibility of the fabrication including material characteristics at high operating temperatures will need to be determined.

CONCLUSIONS AND FURTHER WORK

Adequate heat transfer is an important to the plant configurations considered in the Los Alamos Zero Emission Coal studies. The computer code discussed here is one of the important codes used in the study to help determine the ultimate feasibility of the plant design. In this study the code supplied important information to help with future plant simulations.

This computer code still needs some modifications and improvements that will appear in a second version. The first improvement needed is further automation to reduce the amount of human interface required to get an adequate solution. Another improvement scheduled is to account for entrance effects in computing the heat transfer coefficients for the short tubes used in the fuel cell.

REFERENCES

1. Ziock H.-J., K. S. Lackner and D. P. Harrison *Anerobic Hydrogen Production, Precursor to Zero Emission Coal*. Los Alamos National Laboratory Report LA-UR-00-1850, Los Alamos, NM, April (2000).
2. -----, *Fuel Cell Handbook 5th ed.*, U.S. Department of Energy contract number DE-AM26-99FT40575, Morgantown, West Virginia, October (2000).
3. Minh, N. Q. and T. Takahashi, *Science and Technology of Ceramic Fuel Cells*, Elsevier Science B.V. Amsterdam, The Netherlands, (1995).
4. Kreith, F., *Principles of Heat Transfer*, International Textbook Company, Scranton, PA, (1962).
5. Reid, R. C., J. M. Prausnitz, and T. K. Sherwood, *The Properties of Gases and Liquids, 3ed.*, McGraw-Hill, New York, NY (1977).